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APPLICATION OF THE ACTIVE WELL COINCIDENCE COUNTER TO THE MEASUREMENT OF URANIUM

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Abstract

An Active Well Coincidence Counter has been developed to assay uranium fuel material in field inspection applications. The unit is used to measure bulk UO_2 samples, high enrichment uranium metals, LWR fuel pellets, and ^{233}U-Th fuel materials which have very high gamma-ray backgrounds.

1. Introduction

IAEA inspectors have found the portable High-Level Neutron Coincidence Counter (HLNCC) unit[1,2] particularly useful in field applications for plutonium assay. This instrument can not be used for passive assay of either ^{235}U or ^{233}U because of their extremely low spontaneous fission yields. To make this type instrument applicable to the assay of uranium, we have modified the HLNCC with special end plugs containing AmLi neutron sources to make an active interrogation system that can be used for the verification of high enrichment uranium samples. The modified HLNCC unit has limited sensitivity but can be used to measure metal samples containing several kilograms of ^{235}U.

An improved version of this active system, called the Active Well Coincidence Counter (AWCC),[3] has been built. In comparison with the conventional fast random driver (RD),[4] the AWCC is more portable, light-weight, stable, and less subject to gamma-ray backgrounds. This last feature makes it applicable to ^{233}U-Th fuel cycle materials which generally have very high gamma-ray backgrounds from the decay of ^{232}U. Because the efficiency of the AWCC is about three times higher than the HLNCC, the coincidence counting rate is about an order of magnitude higher for the AWCC. The AWCC can also be used for the passive assay of plutonium by removing the small ($\sim 5 \times 10^4$ n/s) AmLi neutron sources.

In the past, when an interrogation source was used with thermal-neutron well coincidence counters, such as the HLNCC, the 30- to 100-μs gate lengths resulted in a pile-up of accidental counts from the random interrogation source (AmLi). To alleviate this problem, we position the AmLi source inside CH_2 shielding to take advantage of the greater transmission of the induced prompt fission neutrons (2-MeV average energy) compared with the AmLi neutrons (400-keV average energy). With this technique, the induced signal-to-interrogation neutron background ratio can be improved by more than an order of magnitude.

Computer calculations using specially modified versions of the Monte Carlo code, MCNP,[5] are being developed to predict AWCC response for sample geometries and fuel categories outside the range of available standards. These calculations are used to reduce the standards requirements in the calibration of the AWCC.

This paper describes the design of the AWCC and its application to various categories of nuclear materials including high-enrichment uranium metals and low-enrichment bulk UO_2 samples. Standard samples of the above materials have been measured using the AWCC.

2. Instrumentation and Methods

We are investigating two versions of active assay instruments: (1) an add-on version that is designed around the HLNCC and (2) an optimized high-sensitivity version.

HLNCC Modified for Active Assay

We designed the add-on version so that IAEA can use its existing HLNCCs to assay uranium, as well as plutonium. This version is built around the standard HLNCC, except that the top and bottom reflector caps are replaced with source assemblies containing AmLi neutron sources, as shown in Fig. 1. These sources have neutron yields of $\sim 1 \times 10^4$ n/s each and contain ~ 0.1 Ci of Am.

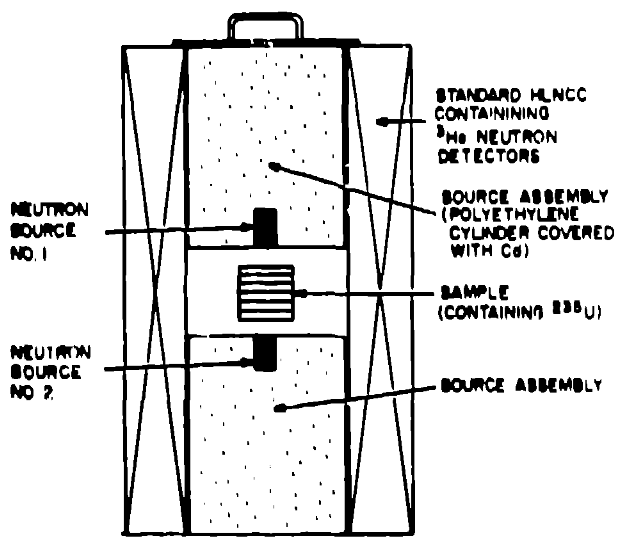

Fig. 1.
Add-on version of the AWCC, which is built around the standard HLNCC.

The sensitivity of this instrument is not high because the neutron detection efficiency of the HLNCC is low (about 11%); however, the instrument is useful for assaying highly-enriched uranium metal samples having masses in the range 1-4 kg. For example, a 3% counting precision (1σ) is obtained on a 2-kg sample in 1000 s.

The useful features of the add-on version are:

1. it is portable,
2. it gives reasonable assay times for highly-enriched metal samples having masses in the range 1-4 kg,
3. the neutron interrogation gives good penetration into the interiors of dense samples, and
4. since the IAEA has several HLNCCs, the addition of the source assemblies make these instruments capable of assaying highly-enriched uranium metal.

Because the performance of such active interrogation systems is proportional to the square of the neutron detection efficiency, significant gains in the performance of such instruments are obtained if the neutron detection efficiency is increased. The AWCC, which is an optimized version of an active assay instrument, is based on a neutron detector having high detection efficiency (~30%). This instrument is described in the next section.

The Active Well Coincidence Counter (AWCC)

Figure 2 is a schematic of the AWCC whose design is optimized for counting the induced-fission reactions and discriminating against the lower energy AmLi background neutrons. The CH_2 moderator and cadmium sleeves are designed for most efficient counting of the induced-fission spectrum neutrons but inefficient counting of the (α,n) neutrons from the AmLi interrogation source.

The nickel reflector on the interrogation cavity wall gives a more penetrating neutron irradiation and a slightly better statistical precision. With the nickel in place, the maximum sample diameter is 17 cm. For larger samples, the nickel can be removed to give a sample cavity diameter of 22 cm.

A cadmium sleeve is placed on the outside of the detector to reduce the background rate from low-energy neutrons in the room. There is also a cadmium sleeve in the detector well to remove thermal neutrons from the interrogation flux and to improve the shielding between the ^{3}He detectors and the AmLi source.

The end plugs have removable CH_2 discs that serve as spacers and can be removed to increase the sample chamber height. Removing the 2.54-cm-thick discs on the top and bottom plugs allows the cavity to accommodate a sample that is 25 cm tall. Larger sizes can be accommodated by removing more discs, as

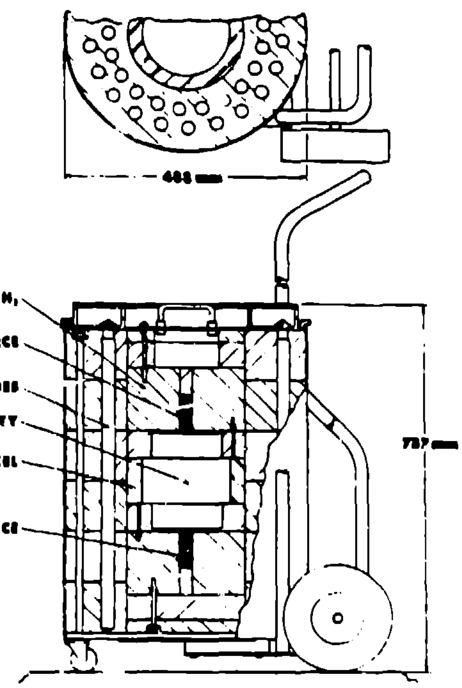

Fig. 2.
AWCC designed for portable use by IAEA inspectors.

shown in Table I. The CH_2 spacers give a higher interrogation flux for small samples (20 cm high) by positioning the AmLi sources closer to the sample. The discs are removable to accommodate larger samples when necessary.

Table I lists the efficiency for counting the fission neutrons from the center of the well and the relative assay time with different end plug configurations. As expected, the measurement is most precise when the sample cavity is smallest. Somewhat unexpectedly, the passive counting efficiency increases at the larger cavity configurations, because the larger end plugs shield the ends of the ^{3}He tubes from the fission neutron source.

The AWCC was designed to use the portable electronics package[6] developed for the HLNCC. To keep this initial model as simple as possible, no neutron flux monitor has been incorporated into the present AWCC. Flux monitors often are used with active neutron assay units to correct for neutron self-shielding or for neutron moderation in hydrogenous matrix materials. Operational experience with the present AWCC will be used to evaluate the need for a flux monitor in more advanced models.

The complete AWCC system, including the electronics, HP-97 calculator, and detector, is shown in Fig. 3. The AWCC and cart weigh approximately 125 kg. The cart allows one person to move the unit over minor obstacles such as small steps.

TABLE I

END PLUG CONFIGURATIONS

Plug Configuration	Cavity Height (cm)	Passive Counting Efficiency[a] (%)	Relative Assay Time[b]
Full discs	20	26	1.00
Small discs removed	25	28	1.44
Both discs removed	35	31	3.03

[a] Corresponds to absolute totals efficiency for a spontaneous fission source (^{252}Cf) placed in the center of the sample chamber.

[b] Corresponds to relative measurement time to reach the same precision for the same sample.

Fig. 3.
AWCC system including the shift register electronics package that is interfaced directly into the HP-97 programmable calculator for automated data collection and analyses.

For assay applications, it is desirable to have a uniform response from all locations within the measurement chamber. To help achieve this goal, AmLi sources are located in both the top and bottom end plugs. This symmetric source arrangement gives a more uniform vertical response. The unformity is helped by the fact that the efficiency for counting the induced-fission neutrons is greatest in the central section of the well counter. To measure the spatial response uniformity, a flat disc sample (1-cm-thick) containing 500 g of ^{235}U was measured at increments from the bottom to the top of the chamber, with the AmLi source positioned in both the bottom and top plugs. The induced response vs height is given in Fig. 4. The response is uniform ($\pm$2%) for sample heights between 5 and 18 cm. For a single source located in the bottom, the response changes by more than a factor of 4 over the same height range.

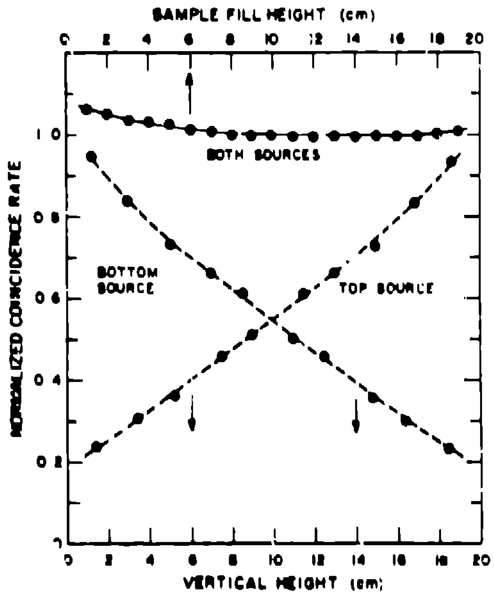

Fig. 4.
Coincidence response as sample fill height for the AWCC in normal configuration using single source (bottom curves) and double source (top curve).

3. Results

High Enrichment Uranium (HEU)

The AWCC has been evaluated for several measurement problems that are of interest to the IAEA. These include: HEU (93% ^{238}U) metal buttons weighing approximately 2 kg, which are input materials to fabrication facilities, and (2) cans of uranium-aluminum scrap generated during manufacture of fuel elements containing tens to hundreds of grams of ^{235}U.

The high enrichment metal discs used in the present experiment are similar to metal buttons of interest in inventory inspections. Two diameters (6- and 7-cm) for the discs were used to check the effect of diameter variations in the measurements. To obtain the mass range from approximately 500-4000 g U, the discs were stacked on top of each other to form a cylinder with heights varying from 1-cm to 7-cm. The uranium metal had a density of 18.7 g/cm^3 resulting in a ^{235}U density of 17.5 g/cm^3. To avoid oxidation and contamination by the uranium, the discs were coated with a thin nickel plate.

Table II gives the results for the different sample masses. The response per gram changes by only 8% in going from 1 disc to 7 discs. There is a cancellation of self-shielding and multiplication effects. For the lower mass region ($<$ 1500 g U) the self-shielding dominates resulting in a decline of the response per g U, but for the higher mass values ($>$ 2000 g U) the multiplication dominates resulting in an increase in the response per g U. The average difference in response per g U from the mean was only 2.7%.

A plot of the coincidence rate versus the response is shown in Fig. 5. The curve is fit through the data points for the 6-cm-diam discs. The 7-cm discs fall slightly above the curve. The average rate per gram for the 7-cm discs is 2.2% higher than for the 6-cm discs.

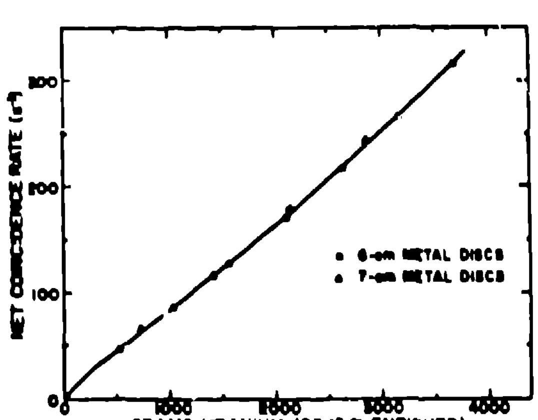

Fig. 5.
AWCC response vs g U for HEU metal buttons.

Low Enrichment Uranium

The AWCC has also been used for the measurement of low enrichment uranium. Because of the low fissile content, the Cd liner is removed and the resulting thermal-neutron interrogation gives increased sensitivity. However, thermal-neutron interrogation is very subject to self-shielding of the neutrons by the uranium resulting in a nonlinear calibration curve.

Materials that normally contain large quantities of hydrogenous materials should be measured with the AWCC in the thermal-mode because the sample matrix material thermalizes the interrogation neutrons. If the AWCC is in the fast-neutron mode, the assay perturbation from the hydrogenous matrix is large; however, if the AWCC is in the thermal-mode, the additional hydrogen in the sample has less of an effect. Examples of this type material are:

1. Scrap and waste mixed with rubber gloves or plastic bags.
2. Uranyl nitrate with concentrations ranging from a few grams per liter to a few hundred grams per liter.
3. Plutonium solutions in the same concen-
tration range as above.

This last category involving Pu solutions would require both a passive (i.e., removal of the AmLi source) coincidence count to obtain the ^{240}Pu effective, and an active interrogation to obtain the fissile content.

To evaluate the AWCC in the thermal-mode, measurements were made on a set of cans containing low enriched U_3O_8. The cans had a diameter of 10.5 cm and a height of 12.5 cm, and typically contained 1 kg of U_3O_8 with the ^{235}U enrichment ranging from 0.7-10%.

The results of the measurements are shown in Fig. 6. The net coincidence response is nonlinear as a function of ^{235}U content because of neutron self-shielding in the U_3O_8. The sample containing 16.8 g ^{235}U gave a 2.2% standard deviation for a 1000-s count, and the sensitivity limit is roughly 1 g ^{235}U.

Passive Assay Applications

By removing the AmLi sources from the end plugs in the AWCC, the unit can be used in the passive mode similar to the HLNCC but with much higher efficiency. In this configuration the unit can be used for measuring the spontaneous fission rates from ^{240}Pu and ^{238}U in bulk samples.

Large mass U_3O_8 samples normally require the removal of the Ni liner and the end plug spacers to accommodate the large container sizes. In this more open configuration, the system has a higher counting efficiency as given in Table I. AWCC coincidence counting rate for ^{238}U is 0.6 counts/s-kg and thus a 10-kg ^{238}U sample (low enrichment) would have an assay precision of about 2% in 1000 s.

TABLE II

HEU METAL BUTTONS MEASURED IN AWCC WITH Ni LINER IN WELL

Sample Size diam x ht. (cm)	Sample Mass[a] (g U)	Coincidence Rate (s^{-1})	Standard[b] Deviation (1000 s run)	$\frac{\text{Counts}}{\text{s·g U}}$ x 1000
6 x 1	524	45.72	3.2%	87.2
6 x 2	1055	86.289	1.55	81.8
6 x 3	1583	127.55	1.2%	80.6
6 x 4	2111	170.30	0.85%	80.7
6 x 5	2636	218.73	0.74%	83.0
6 x 6	3164	268.33	0.64%	84.8
6 x 7	3692	318.33	0.55%	86.2
7 x 1	718	66.22	2.3%	92.2
7 x 2	1434	116.19	1.4%	81.0
7 x 3	2152	179.11	0.92%	83.2
7 x 4	2870	246.65	0.69%	85.9

[a] Uranium metal samples 93.17% enriched in ^{235}U.

[b] Corresponds to relative error in net coincidence rate considering only counting statistics.

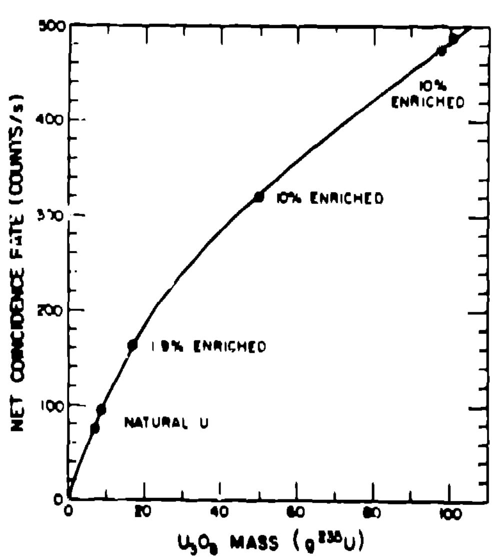

Fig. 6.
Coincidence rate vs ^{235}U mass for low enrichment U_3O_8 samples and thermal-neutron interrogation mode.

To check this application, samples of depleted uranium ranging in mass from 1 to 5 kg were counted in the present detector. The coincidence response was ~ 0.6 counts/s·kg resulting in a statistical error of ~ 2% for a 10-kg sample counted for 1000 s.

The passive application of the detector to plutonium samples is much more important than ^{238}U; however, plutonium applications are described in detail elsewhere[1] for the HLNCC and will not be covered here.

4. Conclusions

The performance characteristics of the AWCC are summarized in Table III. The coincidence counting rates are not highly important because the statistical error is dominated by the accidental coincidence rate. The real coincidence counting rate increases by a factor of 70 in

TABLE III

PERFORMANCE CHARACTERISTICS OF AWCC

	Neutron Interrogation	
	Thermal Mode	Fast Mode
Low-enrichment U_3O_8	11 counts/s g ^{235}U	0.15 counts/s g ^{235}U
High-enrichment metal	NA	0.10 counts/s g ^{235}U
Sensitivity limit[a]	1 g ^{235}U	23 g ^{235}U
Measurement precision (1000 s)	1.5%/20g ^{235}U	3.8%/200g ^{235}U

[a] Defined as net coincidence signal equal to 3 of background for 1000-s count.

going from fast-neutron interrogation to thermal-neutron interrogation. The sensitivity limit is 1 g of ^{235}U for the thermal-neutron mode and 23 g of ^{235}U for the fast-neutron mode. In this case, sensitivity is defined as a net signal equal to three times the standard background deviation over a 1000-s measurement time.

In comparison with the conventional fast random driver (RD),[4] the neutron well coincidence counter is more portable, light-weight, stable, and less subject to effects of matrix materials and gamma-ray backgrounds. This last feature makes it applicable to ^{233}U-Th fuel cycle materials which generally have very high gamma-ray backgrounds from the decay of ^{232}U.

A combination active and passive measurement for plutonium gives both the ^{239}Pu and ^{241}Pu fissile component from the active mode and the ^{240}Pu effective[1] from the passive mode. However, because of the large neutron background for plutonium, larger AmLi sources are needed to override this background in the active mode. This results in assay errors that are larger than can be obtained for ^{235}U.

We have successfully used the system for the assay of HEU metal and UO_2 bulk samples as well as ^{238}U and plutonium.

Future work will include the use of Monte Carlo calculations to extend the range of the measured calibration curves and to include material categories and geometries that are not readily available in the laboratory.

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